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# Grain evolution in conventional and rheo-castings

## A Hellawell

*Department of Metallurgical and Materials Engineering,*
*Michigan Technological University, Houghton, MI 49931, USA*

## Abstract

Nucleation and growth in a conventional, unstirred casting are briefly reviewed and then compared with the probable situation in rheo-cast samples. Emphasis is placed on factors which determine the final grain size within a casting, with particular attention to possible mechanisms of grain multiplication as the microstructure evolves. It is suggested that much of the refinement of the microstructures in rheocastings, may be caused by temperature fluctuations, induced by liquid stirring patterns, rather than by purely mechanical interactions. Metallic dendrites cannot be "broken up" by stirring, but can only be bent elastically or plastically.

## 1. Introduction

There is already a very extensive literature in this field, starting (and continuing) with the earlier papers by Flemings, Mehrabian and coworkers and including some excellent comprehensive reviews (eg. 1-2). The present contribution is solely concerned with the principles involved in grain evolution and the purpose will be to consider mechanisms of nucleation and crystal multiplication as these may affect the final equiaxed grain size in a casting. It is necessary first, to identify possible sources of nucleation and subsequently to consider how these may be influenced by stirring action, mechanical or hydro-magnetic. The approach will be to take what is known about more conventional castings and to see how matters might be modified by the conditions which operate during the production of a rheo-casting. It seems timely to do this before the modelling community meddle.

It must be accepted that all nucleation will be heterogeneous, at relatively small undercoolings, and to distinguish between clean melts and those to which substrates have been deliberately introduced as grain refiners. In the latter case, the nucleation sites are identifiable (although the source of substrate potency may be obscure), while in the former case, nucleation must occur on mould walls or during pouring and gating operations. With subsequent growth, there is then the possibility that crystals may multiply, as by fragmentation of dendrites, whether by ripening, or by mechanical deformation from fluid flow or direct collision. It is important to remark that in metallic systems, close to the melting point, the solid is entirely ductile and dendrites can be bent elastically or plastically, but not fractured. However a plastically deformed dendrite must develop into more than one grain, so that this would, in effect, be a mechanism of crystal multiplication.

In the following we consider, in section 2 nucleation and multiplication in conventional castings, and in section 3, possible extension to rheocastings. A brief summary follows in section 4.

## 2. Nucleation and crystal multiplication

### 2a. Using grain refiners

This is a normal and necessary application in aluminium base castings, using Al-Ti-B combinations (eg. 3). Exactly which substrate surface nucleates aluminium need not concern us here, although it seems probable that nucleation is promoted by a layer of $Al_3Ti$ on a finely dispersed substrate of $TiB_2$ (eg. 4, 5). It appears that in unstirred DC castings, such additions can produce a minimum grain size around $100\mu m$, i.e. $\approx 10^{12}$ grains $m^{-3}$. The reason for there being a maximum attainable grain density, i.e. number of successful nucleation events, can be understood by considering the initial form of a cooling curve, attending recalescence, Fig. 1 (6). At a given cooling rate, as the temperature falls below the liquidus temperature, the nucleation rate rises exponentially. Simultaneously, with cooling, growth beings on each previously nucleated family of substrate particles and the evolution
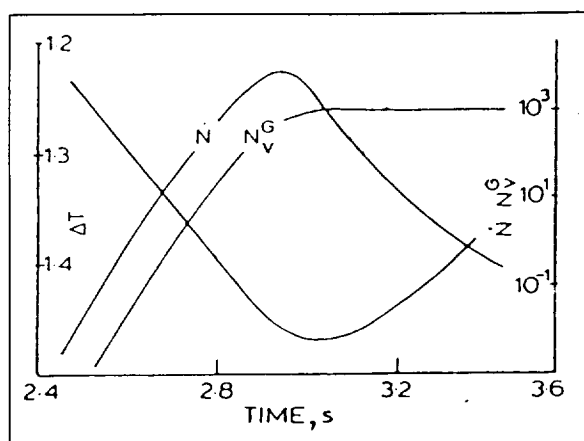
**Figure 1:** Typical cooling curve showing nucleation rate, N, and total number of nucleation events, $N^{G/V}$, during recalescence interval (6).
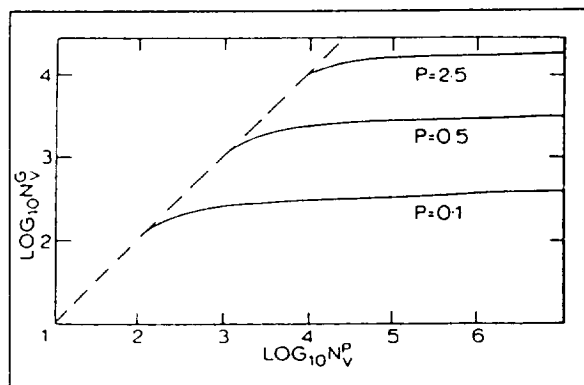


**Figure 2:** Plot of number of grains, $N^{G/V}$, vs. number of dispersed substrate particles, $N^{P/V}$, for three cooling rates, P, in $Ks^{-1}$, assuming one possible nucleation event per particle (6).
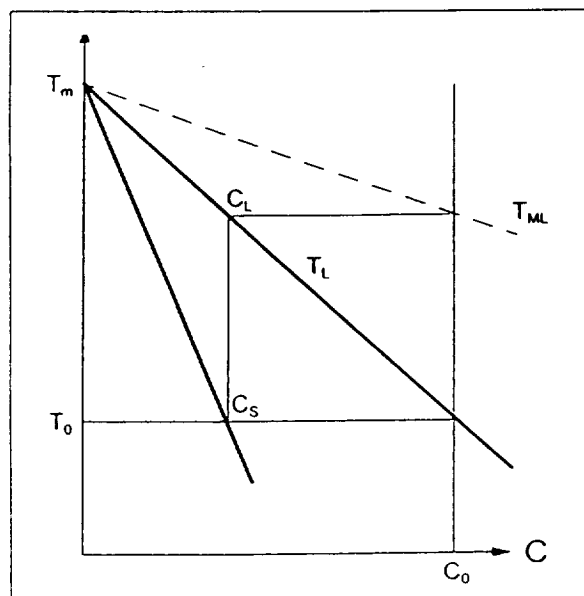


**Figure 3:** A phase diagram for which small particles of solid $C_s$ are initially dispersed in a large volume of liquid, $C_o$, at $T_o$. Melting without composition change occurs at $T_{ML}$, where the solid is in equilibrium with liquid of composition $C_L$ (8).

of latent heat retards the cooling until a minimum temperature is reached, thereafter the temperature rises again to some ambient growth temperature – this is recalescence. The nucleation rate per unit area of available substrate reaches a maximum at the minimum temperature and then falls effectively to zero as the temperature rises, Fig. 1. By the time that recalescence is complete, all nucleation has ceased and the initial/final grain density is predetermined. Any residue of substrate particles which were not involved in this brief interval will remain inactive within the final structure. Therefore, depending upon the magnitude of the maximum undercooling and the duration of the recalescence interval, which are determined by growth kinetics, there must always be a maximum useful addition of grain refiner beyond which there is nothing to be gained. Fig. 2 illustrates this effect for three cooling rates in a given hypothetical system, in which there is a given solute undercooling and growth rate, controlling the minimum temperature attained during recalescence. The plot in Fig. 2 is of number of events (grains), $N^{G/V}$, vs. number of substrate particles, $N^{P/V}$, and assumes a single possible nucleation event per particle. The useful, attainable limit at any cooling rate is where the values of $N^{G/V}$ flatten out. It may be anticipated that this principle will operate in any system and in any type of casting operation, but see later, section 3a.

## 2b. Clean melts

Assuming that melts are "clean" in the sense that they contain no foreign particles which have the potency to nucleate crystals at small undercoolings, nucleation must be confined to mould surfaces, including those of pouring and gating arrangements and of free liquid surfaces (also including bubbles).

With very small superheats, we know that this results in what Chalmers called "big bang" nucleation (7). It is not immediately clear that there is any crystal multiplication involved in this process, but many crystals nucleate on chilled surfaces from which they are swept off into the melt. Whether they are swept off entirely as individual single crystals or as multiple dendritic fragments is an open question; the time frame is very small.

At larger superheats, there are the familiar chill, columnar and equiaxed zones. In a "clean" melt, unless any earlier fragments survived, the equiaxed grains can originate only from within the developing columnar region. It will be necessary to consider how dendrites can fragment within this mushy region and also, how such fragments can escape into open liquid, possibly to survive and grow into equiaxed crystals. Before this, some remarks are appropriate about the probability that particles will survive in a hostile environment above the liquidus temperature.

In the phase diagram of Fig. 3, consider small (eg. 10$\mu$m) solid particle(s) in equilibrium with a large liquid volume with composition $C_o$, at $T_o$. If this solid, of composition $C_s$ is transported into warmer liquid it begins to melt. Disregarding solid state diffusion (life times are brief, < 10s) the melting
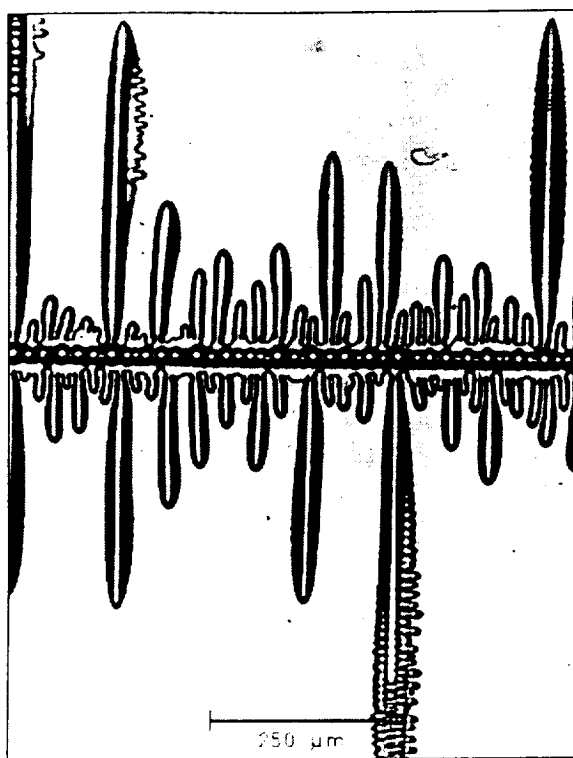
**Figure 4:** A ripened dendrite of $NH_4Cl$, grown from an aqueous solution at steady state conditions, showing how secondary arms are necked at the roots where they join the primary spine, but do not detach (8, 9).

rate is initially controlled by solvent diffusion into the liquid, the rate of which rises with temperature as the composition gradient increases. Simple diffusion expressions allow for crude estimates of the times for such dissolution. However, there is a critical temperature or superheat at which the solid will dissolve without necessary diffusion in the liquid, when melting becomes purely heat transfer controlled. This has been termed a "thermo-solutal" melting temperature (8) and corresponds to the liquidus temperature where the initial solid of composition $C_s$ and liquid, $C_L$, have the same composition, see Fig. 3. Above this temperature the melting rate increases rapidly by some two orders of magnitude and the survival probability effectively falls to zero. The locus of this transition temperature may be read off the phase diagram as $T_{ML}$ and may explain why a high pouring temperature, above this limit, will rarely permit the survival of any solid fragments – this is the opposite extreme to the "big bang" scenario.

We note, that with pre-existing crystal fragments, introduced into a melt as it cools, there is no nucleation barrier, per se, but that there will still be recalescence as growth begins to accelerate during cooling below the liquidus temperature. Recalescence is dominated by time (temperature) dependent growth kinetics.

## 2c. Dendrite fragmentation and fluid flow

Dendrites fragment by local melting at the base of secondary arms, where these join the primary stalk of a dendrite. Fig. 4. These junctions rapidly become necked with ripening

because of local excess in solute rejection from the thickening of primary and secondary branches. Under steady state conditions (rate and gradient) these necked regions are delicately stabilized by the local negative curvature and side arms rarely detach (8,9). Detachment, when it occurs, takes place by local melting at these positions, induced by decreasing growth and/or temperature gradients, or by a local rise in temperature. In the context of constrained columnar, dendritic growth, the rate and gradient are always decreasing more rapidly than the corresponding adjustment of primary dendrite spacing and this promotes detachment by interdendritic solute enrichment. If there is fluid flow in the mushy region, as by thermo-solutal convection (10,11,12), detached side arm fragments may then be ejected out into open liquid, hence to survive or melt, subject to the ambient temperature, c.f. above.

Interdendritic fluid flow probably does not contribute significantly to this fragmentation process, at least in the context of a conventional casting, with/without forced stirring, and is important only as a transportation mechanism. As previously noted, metallic dendrites cannot fracture, only plastic deformation is to be expected and a simple calculation shows that even this is improbable within expected rates of interdendritic fluid flow (13).

Consider, Fig. 5, a schematic side arm cylinder of length L, and diameter d, with a narrower root diameter $d_r$, subject to normal fluid flow at velocity V. The force F, acting on this cylinder is obtained by the same method as that used in the derivation of Stoke's equation (eg. 14, 15) and is equated with the bending moment of the section. Analysis shows that the tensile stress increases parabolically with distance from the
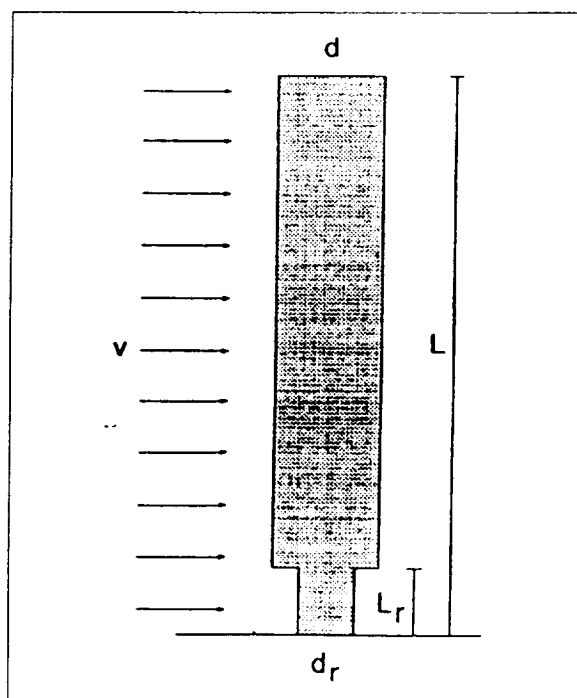


**Figure 5:** Schematic to calculate the bending moment on a cylinder of length L, diameter d and root diameter $d_r$, in a fluid flowing normally at velocity v (13).
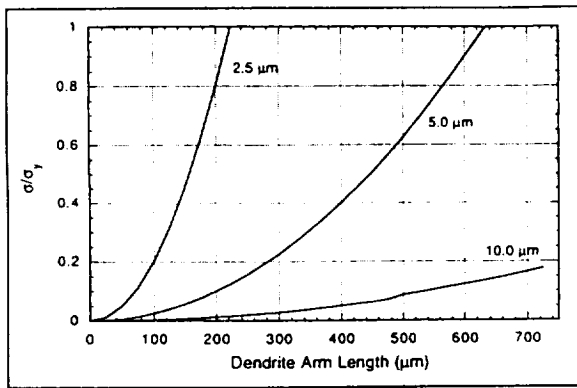
**Figure 6:** Calculated stress, σ, in a dendrite side arm, as a proportion of the yield stress, σy, as a function of the length, L, for three root diameters of 2.5 μm, 5 μm and 10 μm. Refer to Fig. 5 (13).
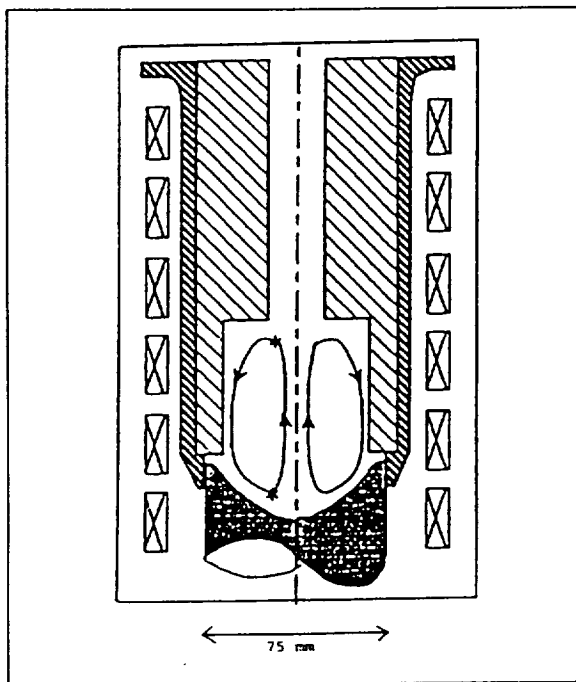


**Figure 7:** Configuration of billet rheocasting equipment from Pechiney (18), showing the pattern of stirring induced by electromagnetic field.

far end of the dendrite side arm until the narrower root, where the tensile stress jumps abruptly as the cross section decreases.

In a typical casting, the main diameter might vary from 10μm to 25μm and root diameters from 5μm to 10μm, while interdendritic flow rates will rarely exceed $10^{-2}ms^{-1}$. With a liquid viscosity of $\approx 3.10^{-3}$ kg.m$^{-1}$s$^{-1}$, the Reynolds number is fairly small ($\approx 0.2$), so that the fluid flow is approximately streamlined. Taking such data, with minimum estimates of the yield stress for aluminium at the melting point, σy, (16), allows comparison of the maximum tensile stress at the root of a side arm, σ, to be expressed as a fraction of this yield stress. In Fig. 6, are plotted σ/σy vs dendrite side arm length, L, for various root radii, dr, for an assumed flow rate, V = $10^{-2}ms^{-1}$, and it is concluded that bending deflections will

nearly always be elastic and rarely reach the plastic level. Refinements of this analysis to include the Reynolds number (17) or with consideration of an array of side arms, rather than a single arm, reinforce this conclusion, namely, that interdendritic fluid flow is unlikely to cause plastic bending of the crystals and therefore that the detachment of side arms is essentially a local melting phenomenon.

# 3. Rheocasting

From the preceding we may consider how the situation might be modified as a melt is vigorously stirred while it cools, using mechanical or hydromagnetic agitation, such as in the schematic of Fig. 7 (18). The solid fraction rises from zero at the top of the assembly to produce a slurry at the bottom having a solid fraction up to 0.8, the local shear rates are high and the mean cooling rates are as high as 0.5 Ks$^{-1}$. The questions arising are what changes might be expected to (a), the nucleation kinetics and (b), the growth morphology and possible crystal multiplication mechanisms?

It is necessary to remark, that with vigorous stirring, especially with hydromagnetic coil configurations producing toroidal movement, local liquid and included solid volume elements are unlikely to cool smoothly as they pass down a column and may well fluctuate across any given isotherm, ie. the local cooling curve will not be smooth, Fig. 8.

## 3a. Nucleation

With heterogeneous grain refining additions to the melt it seems improbable that there should be any significant change in the substrate potency from that occurring in a relatively quiescent melt at a constant cooling rate. The sizes of critical embryos (eg. 10nm) will be far below the thickness of any boundary layer around moving particles. The particles are so small (eg. < 1μm) that their velocities, relative to
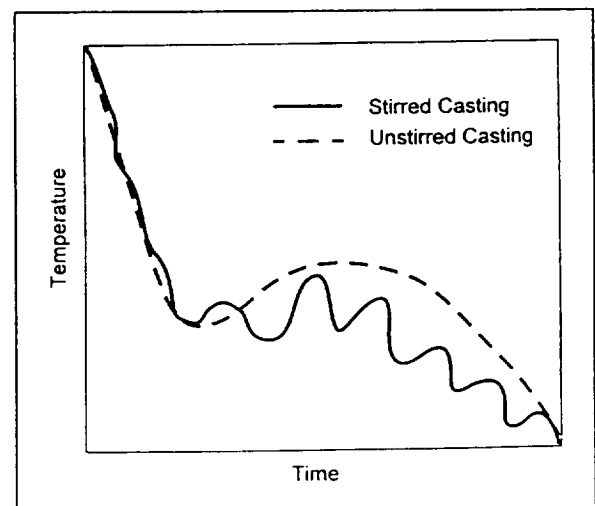


**Figure 8:** Schematic cooling curve, full line, which might be expected for a volume element of liquid which is circulating in a temperature gradient, superimposed on that for quiescent conditions, broken line.
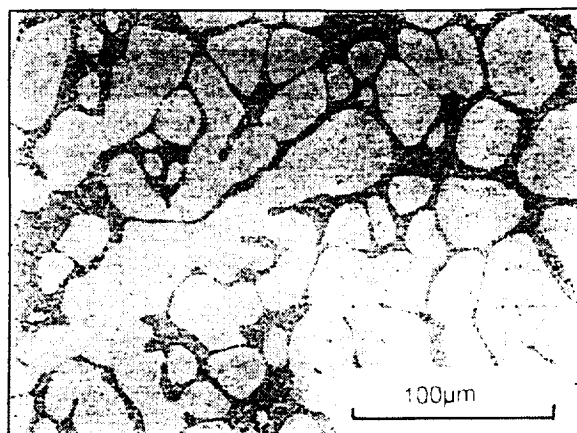
**Figure 9:** Microstructure of rheocast billet of Al-6%Si-3%Mg, produced in the Pechiney assembly of Fig. 7. Some semblance of a dendritic grain stucture is evident.
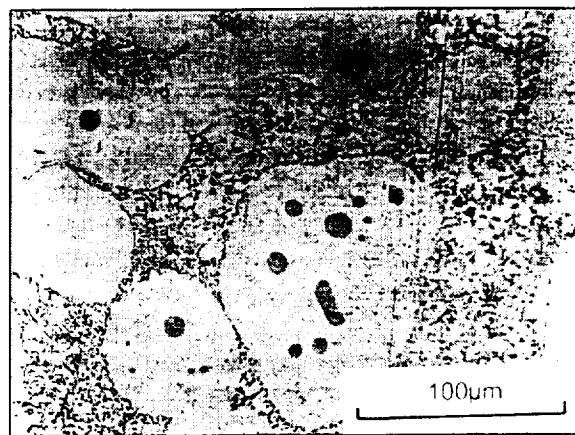


**Figure 10:** Microstructure of the same material as of Fig. 9 after reheating for ≈ 30s just above the eutectic temperature prior to thixo casting. The primary (Al) appears to have coarsened and shapes have become more nearly spherical.

surrounding liquid will be very slow and they will be swept along with the liquid, so that a stagnant boundary layer of thickness comparable with their widths might be expected, ie., far exceeding the scale of nucleation events upon the substrate surfaces. The probability that the cooling path is erratic, however, may alter the eventual number of nucleation events, because there is unlikely to be any clearly identifiable recalescence, but rather an erratic rounding off of the cooling curve with continuous fluctuations in the local temperature, as suggested in Fig. 8. This could allow for more continuous nucleation events. One notes that grain sizes down to ≈ 30μm have been recorded in some rheocastings (2), compared to the limit of around 100μm in unstirred D.C. ingots, or an order of magnitude increase in the grain density. It is possible that such refinement could originate from continuous nucleation in the absence of recalescence.

In a clean melt, only the containing and/or moving surfaces are available as substrates for nucleation. As mentioned previously, we do not know if a crystal which nucleates on such a surface becomes detached as a whole, or whether it adheres to that surface and begins to grow as very fine dendrites which *melt* into fragments, which are then swept away into the open liquid. If this were the case, relatively few such events could act as continuous multiplication sources for crystal fragments, but there is no obvious way to assess this possibility in a quantitative manner.

### 3b. Growth with stirring

The first study of how microstructure evolves during vigorous stirring was carried out by Joly et al. (19), in which Pb-Sn samples were quenched in a counter-rotatory viscometer after various times with increasing solid fractions. The cooling rates were low (eg. 0.01 Ks⁻¹) and isothermal stirring took place over long time periods (over 30 minutes). Clearly, these cooling conditions were much slower and more attenuated than those prevailing in current production of billet stock, as in Fig. 7, but they do provide a starting basis for discussion. As shown, although the initial growth, at small solid fractions, is

probably dendritic, the grains gradually loose this geometrical identity and become smoother, with larger solid fractions than in an unstirred melt. There are no identifiable chill or columnar zones, so that the structure, in conventional terms, is all "equiaxed".

If there is to be impingement and segregation, with or without plastic deformation, it would be expected while the crystals are still relatively open and dendritic. But growth is taking place into vigorously stirred or sheared liquid (although the solid must be swept along with that liquid) and no stable diffusion fields can be established to allow for continued dendrite evolution – so that the smooth, rounded shapes are to be expected, as crystals slide past one another, lubricated by the liquid and making "soft" rather than "hard" contact. One notices that individual solids do show possible evidence of what might be grain boundaries, suggesting that they may be polycrystalline, possibly from earlier collisions, but at longer times these are less obvious and may have been removed by grain growth. The time periods were long in these experiments, as previously noted, and the microstructures in more rapidly produced billet stock, using hydromagnetic stirring are rather different.

Fig. 9 is from an Al-6% Si – 3% Mg rheocast billet, produced in the Pechiney equipment (18, 20) shown in Fig. 7. There are remnants of a dendritic structure, perhaps much clearly defined at the edge of the casting, where there is a thin (≈ 0.1mm) skin of a chill or columnar structure. It is not possible, from a two dimensional section, to know how interconnected the globular shapes might be, except where a dendrite frame is evident, and even then one does not know if side arms are detached from primary spines. Definition of "grain" size is not quite obvious and some orientation studies would be instructive, as from electron channelling maps (21). One might suppose that this structure, which develops within the order of a few minutes, is primarily a consequence of dendrite fragmentation as the fluid is swept in a toroidal pattern, vertically and horizontally within the sump of the mould. Pluchon et al. suggest that this fragmentation is most

probable at the basal sides of the mould where dendritic growth develops. From the previous discussion, c.f section 2c, it might be concluded that the fluid flow does not actually "break off" dendrite fragments, but is responsible for sweeping away those side arms which *melt* off the fine primary spines. If this were the correct interpretation, as with natural convection in an unstirred casting, the role of fluid flow is as a transport mechanism and it does not cause fracture, per se. As noted, previously, metallic dendrites can only be expected to yield elastically or plastically.

Finally, Fig. 10, it is interesting to look at the microstructure after reheating and thixo-casting/forging. The billet stock is typically heated to just above the eutectic temperature, where it is held for less than a minute, prior to injection into the mould. The primary structure has dramatically coarsened and the solid shapes show no trace of orientation. There are numerous included liquid pockets. One assumes that these are indeed, separate globular crystals, but it is puzzling that the structure could ripen and coarsen so rapidly, unless, perhaps, the original and finer shapes of Figs. 9 were to some extent interconnected – i.e. it may be that during the reheat, there is only spheroidization, but not significant coarsening. Some studies of the microstructure with varied holding times would be useful to clarify this development.

## 4. Summary and conclusions

The preceding is a brief commentary on some possible reasons for differences between conventional and rheo- or thixo-castings. The development of this process(es) has taken place very rapidly, often by trial and error, so that an understanding of the physical metallurgy involved is still intuitive, at best. Some suggestions/conclusions might be as follows:

(a) The thermal cooling conditions for a volume element which prevail during the production of a stirred rheocast billet, must involve continuous fluctuations of variable amplitude and duration. It is suggested that this could be a factor which is as important, or perhaps more so, than the purely mechanical consequences of the stirring. Such temperature fluctuations might influence the nucleation kinetics because there would be no clear recalescence and would be expected to promote dendrite fragmentation by local remelting.

(b) In many structures, there is a lack of experimental information about what exactly constitutes a "grain", in the conventional sense. There would appear to be some scope here for detailed orientation studies such as local electron channelling maps might reveal.

(c) If there are to be attempts to model the evolution of grain or particle size, through the rheocasting and remelting operations, it will presently be impossible to do so in a physically predictive way. Presumably all that can be done, without a better understanding, would be to fit the end results by introducing some arbitrary "multiplication coefficient(s)", into a program.

## 6. References

1. 1991 FLEMINGS, MC, Metall. Trans., Vol. 22A, pp. 957-981.

2. 1994 KIRKWOOD, DH, International Materials Reviews, Vol. 39(5), pp. 173-189.

3. 1968 GLASSON, EL, and Emley EF, The Solidification of Metals, Iron and Steel Inst., London, Book No 110, pp. 1-9.

4. 1979 HELLAWELL, A, The Solidification and Casting of Metals, The Metals Soc., Book No. 192, pp. 161-168.

5. 1996 GREER, AL, Metall. and Mater. Trans., Vol 27A (3), pp. 549-556.

6. 1975 MAXWELL, I, and Hellawell A, Acta Metall., Vol 23, pp. 229-239.

7. 1963 CHALMERS, B, J. Australian Inst. Metals, Vol. 8, pp. 225-238.

8. 1996 HANSEN GC, Hellawell A, Lu SZ and Steube RS, Metall. & Mater. Trans., Vol. 27(3), pp. 569-582.

9. 1995 HELLAWELL A, in Modeling of Casting, Welding and Advanced Solidification Processes, Vol. VII, Eds Cross and Campbell, TMS, pp. 565-576.

10. 1970 MCDONALD, RJ, and Hunt JD, Metall. Trans., Vol IA, pp. 1787-1788.

11. 1986 MOORE, JJ, and Shah NA, International Metallurgical Reviews, Vol 28, pp. 287-310.

12. 1993 HELLAWELL, A, Sarazin JR and Steube RS, Phil. Trans. Roy. Soc. London, Vol. 345A, pp. 507-544.

13. 1996 PILLING J, and Hellawell A, Metall. & Meter. Trans., Vol 27A(1), pp. 229-232.

14. 1973 GEIGER, GH and Poirier DR, Transport Phenomena in Metallurgy, Addison-Wesley Pub. 10, Chapters 2 and 3.

15. 1992 GASKELL, DR, An Introduction to Transport Phenomena in Materials Engineering, Macmillan Pub. Co., Chapters 3 and 4.

16. 1982 FROST, HJ and Ashby MF, Deformation Mechanism Maps, Pergamon Press, p. 21.

17. 1967 BATCHELOR, GK, Introduction to Fluid Mechanics, Cambridge Univ. Press, Chapter 4, pp. 244-246.

18. 1995 PLUCHON, C, Loué W, Menet P-Y, and Garat M, Symposium on Processing of Semi-Solid Materials, TMS, Materials Week, Cincinnati, Ohio, and the present proceedings.

19. 1976 JOLY, PA, and Mehrebian R, J. Materials Science, Vol. 1, pp. 1393-1418.

20. 1996 SARAZIN, JR. Hot Metal Moldings, Arkadelphia, Arkansas – Communication.

21. 1988 RANDLE, V, Ralph B and Dingley D, Acta Metall., Vol. 36, pp. 267-280.